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Predictors of Residential Ozone in Danish residences

Elvira V. Bräuner¹, Marie Frederiksen¹, Barbara Kolarik¹, Dorina G. Karotki², Michal Spilak¹, Zorana J. Andersen³, Anni Vibenholt⁴, Thomas Ellermann⁵, Steffen Loft² and Lars Gunnarsen¹

¹Danish Building Research Institute, Aalborg University, Denmark

²Department Public Health, Section of Environmental Health, Faculty of Health Sciences, Copenhagen University, Copenhagen, Denmark

³Department Public Health, Center for Epidemiology and Screening, Faculty of Health Sciences, Copenhagen University, Denmark

⁴National Research Centre for the Working Environment, Copenhagen, Denmark

⁵National Environment Research Institute, Aarhus University

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Abstract

Ground level ozone is a secondary pollutant formed by a series of sunlight driven reactions involving nitrogen oxides and volatile organic compounds arising largely from traffic and enters households via ventilation and infiltration. The health effects of ozone exposure range from respiratory irritation to increased mortality. A large proportion of studies published on the health effects of ozone have estimated individual exposure from measurements obtained from outdoor monitors. But the use of fixed monitoring sites as a surrogate to estimate personal exposure may not adequately reflect personal exposures. Determining the predictors of indoor ozone may aid in recommendations regarding determination of ozone in epidemiological studies and may also aid in reduction information campaigns issued by local governments.

Our purpose was to identify predictors of indoor ozone and assess how well outdoor street level ozone reflects indoor ozone levels. In this study of 26 homes located in Copenhagen, the capital of Denmark, we investigated air filtration, season, housing characteristics and dimensions, presence of gas appliances, ventilation and air exchange rates, occupant behavior, distance to major roads, indoor environmental variables as well as outdoor ozone levels as potential determinants of indoor ozone by linear regression analyses.

Our results indicate that the season of Spring, homes built before 1900, low floor level, proximity to a major road, upper floor level, air intake, and double glazed windows were positively associated with indoor ozone whilst daily window opening, gas stoves, pets and candle burning were negatively associated. In this study outdoor ozone was not consistently associated with indoor ozone.

Epidemiological studies should consider direct measurements of residential ozone levels.

Keywords - residential ozone, predictors

1. Introduction

Ground level ozone is a secondary pollutant formed by a complex series of sunlight driven reactions involving nitrogen oxides and volatile organic compounds arising largely from traffic⁽¹⁾. This pollutant constitutes one of the major outdoor pollutants in industrialised countries. The effects of short-term exposure to ozone in both human trials⁽²⁻⁴⁾ and animal toxicological studies⁽⁵⁾ have been documented and numerous epidemiological studies have reported associations between ozone and daily mortality⁽⁶⁻⁸⁾.

Ozone enters households via ventilation and infiltration and can be generated from indoor sources, upon entering ozone will quickly be consumed by chemical reactions with organic compounds on indoor surfaces or in air, leading to contamination of air with other reactive- or ultrafine particles⁽⁹⁾. Most epidemiological studies published on the health effects of ozone have estimated individual exposure from measurements obtained from outdoor monitors^(2-8;10-12). But the use of fixed monitoring sites as a surrogate to estimate personal exposure may not adequately reflect personal exposures. Although the European limit for ozone of 180 $\mu\text{g}/\text{m}^3$ ⁽¹³⁾ is only exceeded in Denmark one to two times every year evidence suggests that even this limit may not be stringent enough to protect sensitive subjects from adverse effects⁽¹⁴⁾. People in western countries including Denmark spend 85-90% of their time indoors so indoor exposure to ozone and ozone-reaction products may play an important role⁽¹⁵⁾, thus determining the predictors of indoor ozone levels may aid in reduction information campaigns issued by local governments.

Within the present long-term study a large amount of data was collected on housing characteristics, ventilation and indoor quality as well as outdoor ozone levels measured at a fixed monitoring station. The purpose of this paper was to determine predictors of indoor ozone levels in 26 Danish homes located in Copenhagen, the Capital City of Denmark and assess how well outdoor ozone levels reflect indoor levels.

2. Materials and Methods

2.1 Design

Volunteers aged over 50 years were recruited using notices in local newspapers, activity centers and local supermarkets. We recruited a normal population of 50 healthy non-smokers (6 singles and 22 couples living in 28 apartments) all retired or working from home. One couple resigned and another couple was excluded from analyses due to missing ozone measurements, leaving 26 homes, which we based our ozone predictions on.

The homes were located in residential areas of the Greater Copenhagen City in Denmark within 300 m (10 homes) or 300-1700 m (16 homes) of major roads with more than 70,000 vehicles per 24-h. Sampling weeks were allocated to each of the homes taking into account the periods participants were unavailable, and we measured in two homes concurrently each measurement week. All measurements were completed within a 7-month period starting in November 2010. Participants were asked to continue their normal activities throughout the entire sampling period to ensure the concentrations measured were representative.

2.2 Housing characteristics

Housing characteristics and occupant behaviour were determined using self-administered questionnaires at the beginning of the 28-day study. The environmental questionnaire included information on housing characteristics (construction year, housing dimensions, windows, floor covering), ventilation systems, stove and oven type as well as general occupant behavior (frequency of candle burning, food frying, environmental tobacco smoke (ETS), window opening and presence of pets).

2.3 Residential and street level outdoor ozone

Residential ozone concentrations for each address within each 14-day exposure period were determined using passive ozone measurements (Radiello tubes from SUPELCO, Switzerland). Passive samplers were placed in the bedroom and living room of each home. The passive samplers consisted of an adsorbing cartridge inside a blue light-protective diffusive body with outer dimensions: 60 mm height and 16 mm diameter, within which air diffuses by molecular diffusion. The sampling rate for ozone at 25 °C is 24.6 ml/min. The temperature during each measurement period was logged, and the average was used for temperature correction of the sampling rate. The adsorbing cartridge was formed by a micropore polyethylene tube filled with silica gel coated with 1,2-di(4-pyridyl)ethylene solution (DPE) and closed, at one end, by a PTFE cap. Silica gel ensures the presence of water, necessary to complete ozonolysis. Upon exposure to ozone, acid-catalysed ozonolysis of DPE leads to the production of 4-pyridylaldehyde. Addition of 3-methyl-2-benzothiazolinone hydrazone (MTBH) to the tube yields the corresponding azide, yellow coloured complex, which was measured in a ELIZA plate reader spectrometer at 450 nm. The reaction is specific for ozone; and there was no interference from nitrogen oxides or organic compounds. 4-pyridylaldehyde in MTBH solution was used as a standard for the calibration curves. Individual indoor ozone concentrations were calculated as the median and mean at each address for the whole study period as well as according to season.

Data on street level ozone were provided by the Danish Center for Environment and Energy within the same period for each participant. Measurements form part of the routine Danish Air Quality Monitoring Program and have previously been described⁽¹⁶⁾. In brief, outdoor ozone was

continuously monitored at half hour intervals at a fixed monitoring station located at street level on H.C. Andersen Boulevard, using ultraviolet photometry at 254 nm (Ozone monitors API M400 and 400A). Outdoor street level ozone concentrations were calculated as the median and mean exposure at each address for the whole study period as well as according to season.

2.4 Carbon dioxide (CO₂)

CO₂ levels were monitored continuously at 5 minute intervals in the bedroom of each home using the U12 series of HOBO data loggers (Synotech, Huckelhoven, Germany) and data was continuously transmitted to an external PC. Median and mean CO₂ bedroom levels were calculated for the whole exposure period and according to season.

2.5 Air exchange rate

Air exchange rates for each home were determined within each exposure scenario using the perfluorocarbon tracer gas (PFT) method^(17;18). At the beginning of each exposure period PFT emitters and capillary adsorption tube samplers (CATS) were deployed throughout the dwelling with the exception of uninhabited zones such as cellars, attics and storage rooms. Both the PFT emitters and CATS were placed at a height of 1 to 2 m above floor level and away from any heat or cold source as they function according to temperature. Both CATS were collected and analyzed at the end of each 14 day exposure period. For each home, the air exchange, expressed as air changes per hour, was calculated for the whole period and according to season.

2.6 Statistical methods

Potential predictors of indoor ozone levels were determined by generalized linear models using the GLM procedure in SAS (version 9.2; SAS Institute, Cary, NC). The residuals of the ozone models were randomly distributed after transformation of indoor ozone concentrations by the natural logarithm and all regression analyses were performed using these values. The influence of the explanatory variables on indoor ozone levels were calculated by exponential transformation of the regression estimates and expressed as a percentage change in indoor ozone concentrations. The 24 explanatory variables used in all models are presented in tables 1 and 2 and included filter, season, year of construction, number of rooms, floor level, floor area, carpeting, exhaust systems in bathroom and kitchen, air exchange rate, presence of gas appliances, window type, pets in the home, window opening daily, candle burning, ETS, indoor temperature, indoor relative humidity, PM_{2.5}, distance to closest major road, traffic load at front door and outdoor ozone levels.

The following approaches were used to determine predictors of indoor ozone: *i) univariate regression analyses*, where each explanatory variable was included individually in the model; *ii) a multiple regression approach*, including all explanatory variables with mutual adjustment and *iii) a*

backward stepwise linear regression approach. The significance threshold was $p < 0.05$ in approach *i*) and *ii*) and in the last approach the exclusion of the variables continued until the P value of all the included explanatory variables was below 0.10.

3. Results

The homes had a median floor area of 90.5 m² distributed over a median of four rooms. Almost 50 % of the homes were constructed prior to 1900, were located on the 2nd floor or higher, had double glazed windows with no anti-UV coating, and were not equipped with exhaust systems in either the kitchen or bathroom, whilst only 8 homes had no form of trickle or façade air intake. Carpeting on floors was not common with only 3 homes reporting carpeted floors. Traffic loads at the front door were typically over 500 vehicles/day and almost 40% of homes were located within 300 m of a road with over 70 000 vehicles/day. Occupants typically had no pets, were not exposed to ETS and didn't own gas ovens and stoves, whilst around 50% of the occupants opened windows daily and burnt candles at least three times a week.

Median indoor ozone, temperature, relative humidity, CO₂, air exchange and outdoor street level ozone were 0.97 ppb, 21.7 °C, 34.3 %, 684 ppm, 0.5 h⁻¹ and 18.0 ppb, respectively for the whole period. Median outdoor street level ozone was 18.0 ppb for the whole period and significantly higher during spring compared to winter, whilst median outdoor ambient ozone levels were 30.3 ppb.

The estimates in the multivariate model were mostly in agreement with the univariate model (magnitude, direction and significance), but estimates were somewhat larger with wide confidence limits and for a number of the variables the direction of the estimate changed in the multivariate model. Among the 24 explanatory variables season, year of construction, floor level, number of rooms, window type, air intake, stove type, distance to major road, pets, candle burning and window opening remained in the reduced model. Estimates for the variables remaining in the reduced model all resembled those in the univariate models in which each variable was entered alone except for air intake in which the direction of the estimate changed and this model explained 59% of the variation in indoor ozone levels. Concentrations of indoor ozone in the reduced model increased in the season of Spring and in homes built before 1900, located close to a major road, below the second floor, with either façade or trickle air intake, double glazed windows and according to number of rooms. The presence of a gas stove and pets reduced ozone levels and daily window opening and candle burning more than three times per week were also associated with reduced indoor ozone levels. Outdoor street level ozone was positively correlated with indoor ozone in the univariate model, negatively correlated

in the multivariate model and also negatively correlated in the backward regression model prior to exclusion.

4. Discussion

The season of Spring, homes built before 1900, low floor level, proximity to a major road, floor level, air intake, and double glazed windows were positively associated with indoor ozone whilst daily window opening, gas stoves, pets and candle burning were negatively associated. These characteristics explained almost 60% of the variation in indoor ozone levels. Outdoor ozone was not consistently associated with indoor ozone and neither air filtration nor air exchange rates played significant roles in the variation of indoor ozone in the present study.

Levels of indoor ozone reported in this study are in agreement with indoor personal levels reported in previous studies⁽¹⁹⁻²¹⁾ and low indoor ozone levels can partially be attributed to the short indoor half-life of ozone of 6 to 10 minutes indoors⁽²²⁾, as ozone rapidly reacts with indoor materials and becomes decomposed or deposited^(23;24). Also our measurements were taken primarily in the winter when levels are expected to be lower as ozone is formed by sunlight driven reactions dominated by long range transport from more Southern parts of Europe and levels would thus be expected to be higher in summer^(1;16). The urban background ozone concentrations we report in this study were higher than the street levels measured on the busy H.C. Andersons Boulevard. This trend was expected, as urban background ozone at this height and distance to traffic is less subject reactions with nitrogen oxide and thus better reflecting the average outdoor urban exposure. We also observe increased ozone levels in homes built after 1900 compared to younger homes. This might not be related to the building as such, but rather that the older buildings are closer to busy streets with nitrogen oxide traffic emissions consuming outdoor ozone.

Our findings that spring ozone levels are higher than winter levels are in line with the fact that ozone is formed by sunlight driven reactions and levels would thus be expected to be higher in spring^(1;16).

The presence of combustion related sources to NO and particles would be expected to reduce levels of ozone as ozone would quickly be consumed by NO and chemical reactions with organic compounds on the surface of these particles suspended in air^(9;22). This trend is observed in the homes in the present study with reduced indoor ozone in homes with frequent candle burning compared to homes with less frequent candle burning as well as those with gas stoves. The consistent use of dedicated exhaust fans over gas stoves would also explain the effective removal of ozone. In contrast the ETS was associated with higher indoor ozone levels, which may be due shorter exposures or a larger drive toward opening windows after exposure.

The association between increased air exchange rates and higher indoor ozone levels is expected^(22;25), we can confirm this positive relationship in our univariate and multivariate models. Although air exchange rates in this study do in fact conform to the requirements of the Danish Building Regulation (BR10) of 0.5 air exchanges per hour⁽²⁶⁾, the levels are generally low.

Dwellings located above the first floor were associated with increased levels of indoor ozone, probably due to lower levels of reactive nitrogen oxides and volatile organic compounds necessary for ozone chemistry at this floor level, but this estimate may not be conclusive as the direction of the estimate varied in the univariate and multivariate models. Homes with façade and trickle air intakes were associated with increased ozone compared to those with none as more ozone would infiltrate these homes. In contrast infrequent window opening and double glazed windows were associated with higher levels, indicating that the air tightness of the home envelope may contribute to the general accumulation of indoor pollutants including ozone.

Outdoor street level ozone was not consistently associated with indoor ozone in the present study. Although the association between outdoor and indoor ozone was positive in the univariate models, we observed negative correlation in the multivariate model in which all explanatory variables were included with mutual adjustment and outdoor ozone was excluded in our backward regression. Outdoor ozone is of course the major source of indoor ozone, but both positive⁽²²⁾ and negative associations⁽²⁷⁾ have been reported previously. Indoor environmental variables, occupant habits, house size and construction, other possible indoor ozone sources and importantly the distance of the household to the monitoring station at which outdoor ozone is monitored all play major roles in the correlation between indoor and outdoor ozone levels, for example in this study more than 50% of the dwellings were located more than 2 km away from the outdoor monitoring station and this would affect how well the monitor data would reflect the exposures in these homes. As we illustrate in the present study several other variables seem more important in determining ozone levels indoors than the outdoor monitor levels alone. Many epidemiological studies report adverse effects of ozone based on outdoor monitor data as a surrogate of personal exposure^(2-8;10-12), but this may have considerable uncertainty as other important variables are not accounted for, thus the direct measurement of ozone by the use of personal monitors is recommended.

The limitations of this study include the fact that daily variability in concentrations and possible differences between weekday and weekend are not addressed. Measurements were taken during winter and spring and the associations between indoor ozone may change in summer. Indoor ozone levels were low and may reflect saturation of the passive ozone sampler.

5. Conclusion

Epidemiological studies should consider direct measurements of residential ozone levels or the use of personal ozone monitors.

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